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Anomalous superconductivity and field-induced magnetism in $CeCoIn_5$

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In the heavy fermion superconductor $CeCoIn_5$ ($T_c = 2.3$ K) the critical field is large, anisotropic, and displays hysteresis. The magnitude of the critical-field anisotropy in the a-c plane can be as large as 7 T and depends on orientation. Critical-field measurements in the (110) plane suggest two-dimensional superconductivity, whereas conventional effective-mass anisotropy is observed in the (100) plane. Two distinct field-induced magnetic phases are observed: H_a appears deep in the superconducting phase, while H_b intersects Hc_2 at $T = 1.4$ K and extends well above T_c. These observations suggest the possible realization of a direct transition from ferromagnetism to Fulde-Ferrel-Larkin-Ovchinnikov superconductivity in $CeCoIn_5$.

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The interaction of magnetism and superconductivity is a significant and long-standing problem in condensed-matter physics. Usually, the presence of magnetic order undermines superconductivity, but in heavy fermion materials, superconductivity and magnetism can coexist without deleterious consequences to the superconducting state. These systems provide an opportunity to explore the interaction of magnetic and superconducting order parameters as a function of temperature, pressure, or magnetic field.[1] While antiferromagnetism interacting/coexisting with superconductivity is the case most often considered, examples of ferromagnetism coexisting with superconductivity have been reported recently.[2,3] In heavy fermion superconductors the combination of large initial critical field vs temperature slopes and long mean free paths also potentially allows for the observation of critical fields beyond the Pauli limit and, perhaps, inhomogeneous pairing states.[4,5]

Recently the heavy fermion compound $CeCoIn_5$ was observed to superconduct at 2.3 K, the highest T_c yet reported for a heavy fermion superconductor.[6] Specific-heat and thermal transport studies establish that the superconductivity in this material is unconventional.[7] Because crystallographic anisotropy might play an important role in the properties of this tetragonal material[6] and de Haas–van Alphen measurements reveal a two-dimensional character of the Fermi surface,[8,9] a thorough investigation of the anisotropic critical-field temperature phase diagram was undertaken and is reported in this Communication. We observe not only an upper critical field Hc_2 that varies differently as a function of angle in the (100) and (110) planes, but also the existence of field-induced magnetic phases in both the normal and superconducting states of $CeCoIn_5$.

$CeCoIn_5$ forms in the tetragonal $HoCoGa_5$ crystal structure with lattice constants $a = 4.62$ Å and $c = 7.56$ Å.[6,10] The crystal structure consists of alternating layers of $CeIn_3$ and $CoIn_2$. The crystallographic axes of the flux-grown single crystals used in our experiments were determined by Laue x-ray diffraction. The [001] axis was parallel to the shortest dimension of the crystal and [100] and [010] axes were parallel to the natural edges of the nominally square crystals. The superconducting-normal phase boundary and the magnetization of $CeCoIn_5$ were determined by electrical transport, ac susceptibility, and especially cantilever magnetometery measurements as a function of magnetic field (0–20 T) and temperature (0.020–27 K). Although cantilever measurements can produce some ambiguity in the actual magnetization measured, due, for example, to contributions from both magnetic torque and magnetic force, they are an excellent and unambiguous phase-transition detector. Angular variations were measured using a rotating sample stage[11] in a top-loading dilution refrigerator and in a ^{3}He cryostat. Three different single crystals were studied, with consistent agreement among their measured Hc_2 values.

Figure 1 shows a signal proportional to $\vec{M}$ deduced from cantilever magnetometer measurements plotted against applied magnetic field for both increasing and decreasing fields. The two traces are for the field applied along the [110] and [001] crystal axes at $T = 20$ mK. With increasing field, a narrow superconducting-normal transition, $\Delta Hc_2 < 1$ mT, is clearly seen in the $\vec{H} \| [110]$ trace, and a somewhat broader transition is seen in the $\vec{H} \| [001]$ trace. These traces are typical of data used to construct the phase diagrams reported below. At $T = 20$ mK and $\vec{H} \| [001]$, $Hc_2 = 5.5$ T and for $\vec{H} \| [110]$, $Hc_2 = 11.9$ T. Resistivity measurements (not shown) confirm that these transitions correspond to superconducting-normal transitions. For a material having $T_c = 2.3$ K, these values of Hc_2 are quite large: a simple estimate of the Clogston limit gives Hc_2 $(T=0) = (1.86$ T/K$)T_c = 4.3$ T.[12] For field applied along [110] the normal-superconducting resistive transition occurs at the same field on both upsweep and downsweep; however, the magnetization transition occurs at a lower field on the downsweep, suggesting an additional phase transition in the superconducting state (see Fig. 4 below). No such second transition is observed for $H \| [001]$.

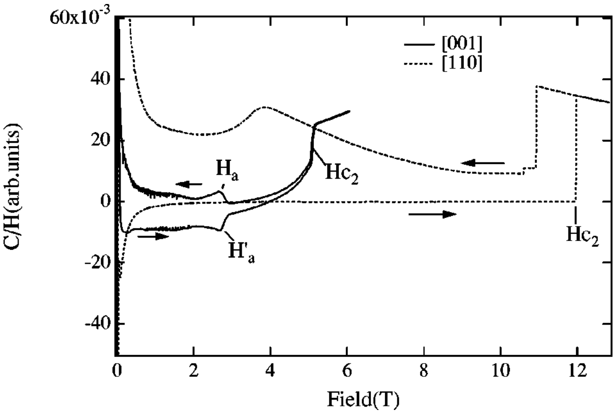

FIG. 1. Magnetization loops for field applied along [110] and [001] at 20 mK in $CeCoIn_5$.The *Y* axis is capacitance/field, which is proportional to magnetization. Arrows indicate the direction of field sweep. The fact that *M* appears to decrease with increasing *H* at high field is likely a result of nonlinear cantilever response rather than an intrinsic negative susceptibility

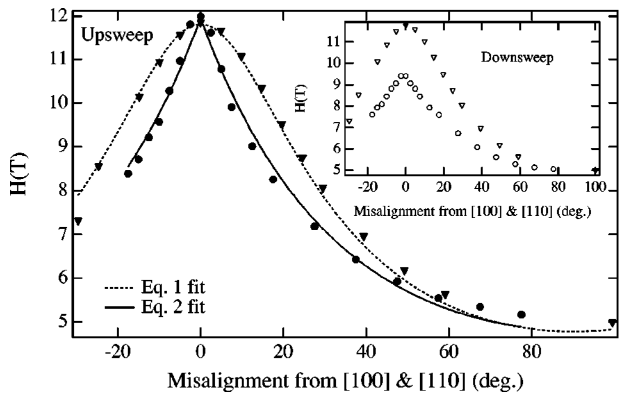

FIG. 2. Hc_2 as a function of angle for $CeCoIn_5$ for field rotations from [110] to [001]=● and from [100] to [001]=▼. Inset shows the down-sweep values for the same rotations: [110] to [001]=○ and [100] to [001]=▽. See text for fit equations.

An additional feature apparent in Fig. 1 is the peak in magnetization observed for $H \| [001]$ at $H_a = 2.8$ T. Preliminary investigations show that this feature appears only below 100 mK and exhibits a complex dependence on field orientation and sweep direction. Although it will be discussed in detail elsewhere,[13] we note here that H_a appears to merge with Hc_2 (upsweep) when the applied field is within 5° of [110].

The angular dependence of Hc_2 at 20 mK is shown in Fig. 2. The evolution of Hc_2 for rotation of $\vec{H}$ from [001] into [100] is well described by the anisotropic effective-mass model,[14] taking $Hc_2(\theta)$ as the up-sweep value:

$$Hc_2(\theta) = Hc_2(\theta=0)/[\cos^2(\theta) + \alpha \sin^2(\theta)]^{1/2}, \qquad (1)$$

where θ is the angle of the applied field out of the tetragonal basal plane and α is the ratio of effective masses $m^*(\theta=0)/m^*(\theta=90)$. The large value of $\alpha=6.1$ confirms the significant electronic anisotropy in $CeCoIn_5$ deduced from de Haas–van Alphen measurements.[8,9]

Rotating $\vec{H}$ from [001] into [110] produces a much more cusplike angular dependence than Eq. (1) would predict. In this case, the data are well described by Tinkham's equation for Hc_2 as a function of angle in thin-film superconductors,[15]

$$|Hc_2(\theta)\sin(\theta)/Hc_2(90)| + [Hc_2(\theta)\cos(\theta)/Hc_2(0)]^2 = 1. \qquad (2)$$

Both sets of data in Fig. 2 were obtained using the same single crystal, so neither sample-to-sample variation nor demagnetization corrections can explain the different angular variations in Hc_2. For these increasing field sweeps going from the superconducting mixed state to the normal magnetic state the demagnitization factors are negligable. We also note that $Hc_2[110] = 11.9$ T while $Hc_2[100] = 11.8$ T which agrees with the reported results of an in-plane anisotropy in Hc_2.[13,16]

The angular dependence of Hc_2 observed in the (110) plane is reminiscent of behaviors in granular thin film[17,18] and multilayer[19] systems. In fact, the quality of the fit to our data is comparable to and extends over a wider angular range than that in Al films.[17] Why two-dimensional (2D) behavior in one particular plane would be observed in bulk $CeCoIn_5$ is not understood. Band-structure calculations suggest that the density of states in the MIn_2 layer in $CeMIn_5$ is quite low[20] and leads to the speculation that perhaps $CeCoIn_5$ may behave as a pseudo $CeIn_3$:$CoIn_2$ multilayer system. Even if such a speculation were shown to be relevant, why the phenomenon would manifest itself in [001]-[110] rotations but not [001]-[100] rotations is unclear; however, it might be related to an in-plane modulation of the superconducting gap function[7,16] or to anisotropic Fermi-surface nesting.[8,9]

The field at which the up-sweep and down-sweep transitions in magnetization occur is a strong function of crystallographic direction (Fig. 2 inset) and these effects are, however, independent of field sweep rate. The difference increases as the field is rotated toward [110] and has a maximum value of 2.5 T. As will be discussed below, $CeCoIn_5$ displays a field-induced magnetic transition at high fields and temperatures above T_c resulting in the presence of a static magnetization in the sample. The values shown in the inset to Fig. 2 have been corrected to account for the magnetization $\vec{M}$ in the sample that contributes to the internal magnetic field $\vec{B}$ according to the relation: $\vec{B} = \vec{H} + \mu_0 \vec{M}$. Using this relation, we corrected an offset in the measured down-sweep transition field around [110] that was the result of moving from a magnetic normal state into a superconducting one. The maximum contribution of $\vec{M}$ is estimated to be $\vec{M} = 1.5$ T/μ_0 along [110]. The essentially zero separation in transition fields as a function of angle for the [100] rotation is shown in the inset to Fig. 2 as well. The maximum field separation along [100] is only 0.08 T, a factor of 31 less than the value of 2.5 T that is found along [110].

The evolution of these up-sweep and down-sweep transitions in the [110] direction with temperature also is anomalous. An *H*-*T* phase diagram for $H \| [110]$ in $CeCoIn_5$ is shown in Fig. 3. Three characteristic temperature ranges can be identified (see Fig. 4 for representative data): (I) For $T < 1.4$ K, field-separated up- and down-sweep transitions in

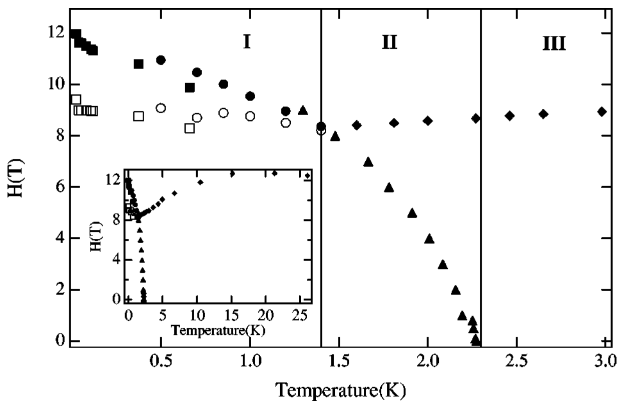

FIG. 3. H-T diagram for $CeCoIn_5$ with $\vec{H}$ applied in the (110) direction (the inset emphasizes the high-temperature range of the main figure). Circles and squares denote magnetization transitions (■ = up sweep, □ = down sweep) and (● = up sweep, ○ = down sweep). Triangles indicate resistively determined Hc_2. Measurements were made with three different systems and the resultant offset between circles and crosses is due to slight differences in crystal alignment. The diamonds denote the field-induced magnetic transition that appears at 1.4 K. I, II, and III indicate the three regions in the phase diagram discussed in the text.

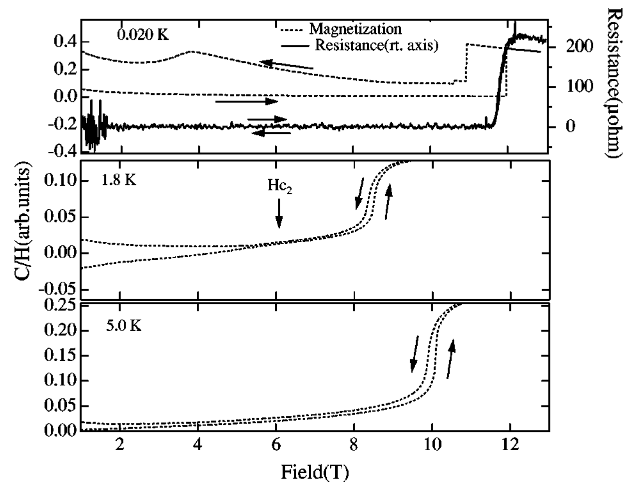

FIG. 4. Magnetization as a function of field ($\vec{H}\|[110]$) at three characteristic temperatures (0.020, 1.8, and 5 K) in $CeCoIn_5$. The Y axis is capacitance/field, which is proportional to magnetization. The noise in the 0.020-K resistance measurement is due to flux exclusion in the magnet at low fields. Note that the sharp transition in the magnetization in the 0.020-K panel occurs at the onset of superconductivity as displayed in the resistance measurement. The arrow indicates position of zero-resistance transition for the 1.8-K panel.

magnetization are observed and the changes in magnetization at both of the transitions are steplike; (II) for $1.4<T<2.3$ K, no evidence for the lower-field transition nor a step in the magnetization at Hc_2 is observed, but for fields greater than Hc_2, a normal-state metamagnetic transition, occurring at H_b, is seen; and (III) for $T>2.3$ K only the field-induced magnetic transition is observed.

In region I the field separation between up- and down-sweep magnetization transitions decreases with increasing temperature and at 0.5 K, $Hc_2(\theta)$ has the same relative angular dependence (not shown) as at 20 mK (Fig. 2). The zero-resistance transition occurs (regardless of field sweep direction) at the higher up-sweep value of Hc_2 deduced from magnetization, and a new ferromagneticlike (because of the observed steps in magnetization) first-order transition in the superconducting state emerges below the resistively determined Hc_2. This state is found below $0.6T_c$ at high fields and its appearance depends strongly on the orientation of field with respect to crystallographic axes. Taken together, these observations are consistent with a spatially inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state.[5] The fact that no signature of a BCS-FFLO transition prior to the superconducting-normal transition is observed in up-sweep magnetization may suggest that this transition is hysteretic in field or that the FFLO state is only stabilized by the presence of magnetic order. High-field heat capacity and neutron-scattering measurements should be able to clarify this issue.

Although the FFLO state is rarely observed,[21,22] $CeCoIn_5$ satisfies the essential conditions for its existence:[23] it is in the clean limit,[7] has a quasi-2D Fermi surface,[8,9] and has an Hc_2 much larger than the Clogston limit. The transition from the normal state to the FFLO state is from ferromagnetic to superconducting. Recent calculations of Zeeman effects in d-wave superconductors[24] (e.g., $CeCoIn_5$, Refs. 7 and 16) suggest that an increase in Hc_2 and the appearance of another magnetic transition, perhaps related to H_a, at lowest temperatures is a consequence of an FFLO state in such a superconductor. If we are not observing FFLO superconductivity in the [110] direction, then the finite jump in $\vec{M}$ in the superconducting state implies the coexistence of superconductivity with a spin-polarized state, the field-sweep dependent continuation of $H_b(T)$ into the mixed state.

The signature for $Hc_2(T)$ intersects $H_b(T)$ and the signature of the FFLO state vanishes at $H=8.0$ T and $T=1.4$ K. Because the magnetization change at Hc_2 disappears above 1.4 K, we used transport measurements to follow Hc_2 up to T_c $(H=0)$ with no observable hysteresis nor second transitions present. The change in magnetization at H_b is approximately a factor of 2.5 less than at Hc_2 (for $T<1.4$ K) and appears to be second order as a function of temperature. The signature for H_b weakens as $\vec{H}$ is rotated away from [110] and is completely absent for $H\|[001]$, again illustrating the anisotropic magnetic behavior of this material even in the normal state. Given the extent to which its evolution is influenced by superconductivity without deleterious effects on Hc_2, it is tempting to identify the paramagnetic-magnetic normal-state transition with spin polarization of a sheet of Fermi surface; quantum oscillation measurements to test this hypothesis are in progress.

In summary, we find a remarkable Hc_2 anisotropy in $CeCoIn_5$ that is correlated with the presence of a magnetic transition in the superconducting state for $H\|[110]$. These data can be described empirically in terms of 2D superconductivity and suggest the formation of an FFLO state. Hc_2 anisotropy also exists within the (100) plane but is describable by anisotropic band-structure effects, and does not present hysteresis. We also have observed two magnetic phases in $CeCoIn_5$, one occurring deep in the superconducting state at very low temperature (H_a) and the other (H_b) manifesting itself as a field-induced metamagnetic transition that persists to at least 25 K.

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[1] N.D. Mathur, F.M. Grosche, S.R. Julian, I.R. Walker, D.M. Freye, R.K.W. Haselwimer, and G.G. Lonzarich, Nature (London) **394**, 39 (1998).

[2] S.S. Saxena, P. Agarwal, K. Ahilan, F.M. Grosche, R.K.W. Haselwimmer, M.J. Steiner, E. Pugh, I.R. Walker, S.R. Julian, P. Monthoux, G.G. Lonzarich, A. Huxley, I. Sheikin, D. Braithewaite, and J. Floquet, Nature (London) **406**, 587 (2000).

[3] C. Pfleiderer, M. Uhlarz, S.M. Hayden, R. Vollmer, H.v. Lohneysen, N.R. Bernhoeft, and G.G. Lonzarich, Nature (London) **412**, 58 (2001).

[4] K. Gloos, R. Modler, H. Schimanski, C.D. Bredl, C. Geibel, F. Steglich, A.I. Buzdin, N. Sato, and T. Komatsubara, Phys. Rev. Lett. **70**, 501 (1993).

[5] Peter Fulde and Richard Ferrel, Phys. Rev. **135**, A550 (1964); A.I. Larkin and Yu.N. Ovchinnikov, Zh. Éksp. Teor. Fiz. **47**, 1136 (1964) [Sov. Phys. JETP **20**, 762 (1965)].

[6] C. Petrovic, P.G. Pagliuso, M.F. Hundley, R. Movshovich, J.L. Sarrao, J.D. Thompson, and Z. Fisk, J. Phys.: Condens. Matter **13**, L337 (2001).

[7] R. Movshovich, M. Jaime, J.D. Thompson, C. Petrovic, Z. Fisk, P.G. Pagliuso, and J.L. Sarrao, Phys. Rev. Lett. **86**, 5152 (2001).

[8] Donavan Hall, E. Palm, T. Murphy, S. Tozer, Z. Fisk, U. Alver, R.G. Goodrich, J.L. Sarrao, and P.G. Pagliuso, Phys. Rev. B **64**, 212508 (2001).

[9] R. Settai, H. Shishido, S. Ikeda, Y. Murakawa, M. Nakashima, D. Aoki, Y. Haga, H. Harima, and Y. Onuki, J. Phys.: Condens. Matter **13**, L627 (2001).

[10] Ya.M. Kalychak, V.I. Zaremba, V.M. Baranyak, V.A. Bruskov, and P.Yu. Zavalii, Russ. Metall. **1**, 213 (1989).

[11] E.C. Palm and T.P. Murphy, Rev. Sci. Instrum. **70**, 237 (1999).

[12] A.M. Clogston, Phys. Rev. Lett. **9**, 266 (1962).

[13] T.P. Murphy *et al.* (unpublished).

[14] L.N. Bulaevskii, Int. J. Mod. Phys. B **4**, 1849 (1990).

[15] M. Tinkham, Phys. Rev. **129**, 2413 (1963).

[16] K. Izawa, H. Yamaguchi, Y. Matsuda, H. Shishido, R. Settai, and Y. Onuki, Phys. Rev. Lett. **87**, 057002 (2001).

[17] Wenhao Wu, R.G. Goodrich, and P.W. Adams, Phys. Rev. B **51**, 1378 (1995).

[18] P.W. Adams, P. Herron, and E.I. Meletis, Phys. Rev. B **58**, R2952 (1998).

[19] F.Y. Ogrin, S.L. Lee, A.D. Hillier, A. Mitchell, and T.H. Shen, Phys. Rev. B **62**, 6021 (2000).

[20] Y. Haga, Y. Inada, H. Harima, K. Oikawa, M. Murakawa, H. Nakawaki, Y. Tokiwa, D. Aoki, H. Shishido, S. Ikeda, N. Watanabe, and Y. Onuki, Phys. Rev. B **63**, 060503(R) (2001).

[21] M.R. Norman, Phys. Rev. Lett. **71**, 3391 (1993).

[22] K. Tenya, S. Yasunami, T. Tayama, H. Amitsuka, T. Sakakibara, M. Hedo, Y. Inada, Y. Haga, E. Yamamoto, and Y. Onuki, Physica B **259-261**, 692 (1999).

[23] J. Singleton, J.A. Symington, M.-S. Nam, A. Ardavan, M. Kurmoo, and P. Day, J. Phys.: Condens. Matter **12**, L641 (2000).

[24] Kun Yang and S.L. Sondhi, Phys. Rev. B **57**, 8566 (1998).